

Photoluminescence from Microcavities Strongly Coupled to Single Quantum Dots

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Abstract

We study theoretically, the photoluminescence properties of a single quantum dot in a microcavity under incoherent excitation. We propose a microscopic quantum statistical approach providing a Lindblad (thus completely positive) description of pumping and decay mechanisms of the quantum dot and of the cavity mode. Our analytical results show that strong coupling (SC) and linewidths are largely independent on the pumping intensity (until saturation effects come into play), in contrast to previous theoretical findings. We shall show the reliable predicting character of our theoretical framework in the analysis of various recent experiments.

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Cavity quantum electrodynamics (CQED) studies the interaction between a quantum emitter and a single radiation-field mode. When an atom is strongly coupled to a cavity mode [1], it is possible to realize important quantum information processing tasks, such as controlled coherent coupling and entanglement of distinguishable quantum systems [2]. In this respect solid-state devices, and in particular semiconductor quantum dots (QD), are the most promising architectures for the possibility of miniaturization, electrical injection, control and scalability. Indeed semiconductor quantum dots provide nanoscale electronic confinement resulting in discrete energy levels, and an atom-like light-matter interaction. Cavity quantum electrodynamics addresses properties of atomlike emitters in cavities and can be divided into a weak and a strong coupling regimes. For weak coupling, the spontaneous emission can be enhanced or reduced compared with its vacuum level by tuning discrete cavity modes in and out of resonance with the emitter. However, when the interaction strength overcomes losses, the system enters the so-called strong coupling (SC) regime [3, 4]. In this case the usual irreversible spontaneous emission dynamics changes into a reversible exchange of energy between the emitter and the cavity mode.

Commonly, these solid state systems in the SC regime are characterized with respect to their behaviour under optical incoherent pump excitation [2, 3, 4, 5, 6]. In this situation, the pump creates an incoherent population of electron-hole pairs typically far above resonance which relaxes incoherently into the QD through various different mechanisms before being emitted by recombination. Moreover, some experimental observations of SC of QDs in microcavities, indicate that the cavity mode is weakly coupled to others various electronic transitions of the system which would contribute in feeding the cavity mode as well, thus resulting in a second incoherent pumping channel. The specific interplay of photon and exciton pumping and decay results in a mixed quantum steady state that influences considerably the observed spectra as we are going to show in detail (see Fig. 1b). An analogous incoherent excitation would also be achieved in the case of current injection, highly desired for the development of optoelectronic quantum devices. Hence, appropriate theoretical modeling of SC with semiconductor QDs under incoherent excitation with a reliable predicting character are sought in order to fulfill the great expectations nowadays attended from future implementations of SC in QD systems. Of particular interest is the analysis for increasing pump excitation while the system is brought into the nonlinear regime.

Recently, Laussy *et al.* [7] reports on the first theoretical analysis of this situation. Their

master equation includes pumping as absorption from incoherently populated reservoirs and spontaneous emission of excitons (radiative recombination) as well as of photons (cavity losses). Their bosonic model is well suited for the investigation of the very low excitation regime where linear optics dominates. It is currently heavily exploited (see e.g. Refs. [5, 6]) to analyze the SC of single QDs as it, at least at first sight, displays an impressive predicting character maintaining a relative easy picture and analytical results. Nevertheless the results obtained within their approach display some puzzling features which deserve careful investigations. As instance, the dependence of the polariton broadenings, of the Rabi splitting and of the spectra on the incoherent pumping rates as well as amplification effects are commonly unexpected results of an harmonic linear dynamics.

The authors have the merit to have pointed out clearly the importance of the effective feeding the other electronic transitions (viz. the other QDs existing in the sample), weakly coupled to the cavity mode. This scenario sets a definite departure from atom-QED models with expected new peculiar features (see e.g. Fig. 1b and Fig. 2b) which are expected to influence even the quantum statistics of the emitted quanta. Anyway, although having a clear and reasonable physical meaning, the master equation of Ref. [7] is not of Lindblad form and then there is no guarantee that it safely generates a completely positive (CP) open dynamics [8]. Actually the obtained photon and exciton populations (see e.g. Eq. (3) of Ref. [7]) can diverge as soon as pumping rates approaches the values of the dampings and even assume negative values as well as it starts to overcome. This highly undesired feature puts into question also the results obtained at lower pumping rates when populations remain finite and positive. The puzzling features, mentioned above, could be the tail (at lower excitation densities) of such problems. Moreover, the need for a fresh, flexible and direct tool for the analysis of state-of-the-art experiments [5, 6] is leading various groups to take advantage of this machinery. These possible artifacts could affect strongly the interpretations of the experimental data and then have negative repercussions on applications and device modeling.

In this letter we provide a theoretical model able to describe SC of a single QD in a semiconductor microcavity under incoherent excitation in the low and intermediate excitation regimes where nonlinear optical effects start to appear. In order to avoid inconsistencies and artifacts, we will start from a microscopic (though simple) description of the pumping and decay mechanisms of the QD and the cavity mode. The picture that we have in mind is

that of two strongly coupled subsystems (the cavity mode and the quantum dot excitations), each in interaction with two independent reservoirs providing both damping and pumping mechanisms. The laser generates electron-hole pairs in the continuum wetting layer which, subsequently, relax into the dot (the P_x pump contribution) by means of incoherent scattering mechanisms such as exciton-exciton scattering mediated by phonons. At the same time, the cavity mode is weakly coupled to others various electronic transitions of the system which would contribute in feeding the cavity mode as well, resulting in a second incoherent pumping channel (P_a). In the limit of weak excitation density the resulting dynamics coincides with that of two strongly coupled harmonic oscillators in the presence of reservoirs. Nevertheless the obtained analytical results significantly differs from those of Ref. [7]. In particular, in the low excitation regime we obtain a very simple analytical expression for the PL spectrum which is definite positive, do not display any amplification effect as well as any change of the Rabi splitting and of the broadenings as a function of the pump intensities. For higher pump densities saturation effects and even lasing effects come into play.

The master equation for this strongly interacting system can be written as

$$\dot{\rho} = i[\rho, H_S] + \mathcal{L}_{MC}^R + \mathcal{L}_{QD}^R, \quad (1)$$

where the system Hamiltonian reads

$$H_S = \omega_a a^\dagger a + \omega_x \sigma_+ \sigma_- + g(a^\dagger \sigma_- + a \sigma_+), \quad (2)$$

with g being the interaction strength between the cavity mode (with annihilation operator a) at energy ω_a and the lowest energy (ω_x) quantum dot exciton with transition operator from the ground state to the exciton level $\sigma_+ = |e\rangle \langle g|$ ($\sigma_- = \sigma_+^\dagger$). The superoperators \mathcal{L}_{MC}^R and \mathcal{L}_{QD}^R describe the interaction of the cavity mode and of the QD with the reservoirs providing both damping and pumping mechanisms. Transmission and diffraction losses of the cavity mode can be modeled, within a quasimode picture, as an effective coupling (γ^c) with an ensemble of electromagnetic modes through the output mirror [9, 10]. When dealing with optical frequencies it can safely be regarded as a zero temperature reservoir [10]. The second mechanism describes the incoherent optical pumping of the cavity. It takes into account that in these samples there may be QDs or more generally electronic transitions weakly coupled to the cavity, in addition to the one that undergoes SC, determining an effective pumping of the cavity mode (P_a). By applying the usual Born-Markov and rotating-wave approximations,

we obtain,

$$\mathcal{L}_{MC}^R = \frac{P_a + \gamma_a}{2}(2a\rho a^\dagger - a^\dagger a \rho - \rho a^\dagger a) + \frac{P_a}{2}(2a^\dagger \rho a - a a^\dagger \rho - \rho a a^\dagger), \quad (3)$$

where $\gamma_a = \gamma^c + \gamma^p$ contains contributions from both the reservoirs and P_a is the total pumping rate depending on the populations of the electronic levels weakly coupled to the cavity mode. Assuming only direct and weakly pumped electronic transitions we obtain $P_a = \sum_i \gamma_i^p \langle n_i \rangle$ and $\gamma^p = \sum_i \gamma_i^p$, where $\langle n_i \rangle$ is the population of the i -th level and $\gamma_i^p = 4\gamma_i g_i^2 / [\gamma_i^2 + (\omega_i - \omega_a)^2]$, being γ_i the inverse of the dephasing time of the i -th transition. The form of the two terms in eq. (3) clearly shows the physical emerging picture different from that addressed in Ref. [7]. Indeed, the cavity experiences radiative spontaneous emission guided by vacuum fluctuations (proportional to γ^c), while the second reservoir provides both emission (proportional to $\gamma_i^p(\langle n_i \rangle + 1)$) and absorption (to $\gamma_i^p \langle n_i \rangle$).

The material excitation strongly coupled to the cavity mode is also under the influence of two different reservoirs: $\mathcal{L}_{QD}^R = \mathcal{L}_{QD}^{se} + \mathcal{L}_{QD}^P$. The first term describes spontaneous emission in all the available light modes except the cavity one,

$$\mathcal{L}_{QD}^{se} = (\gamma_x/2)(2\sigma_- \rho \sigma_+ - \sigma_+ \sigma_- \rho - \rho \sigma_+ \sigma_-). \quad (4)$$

The latter

$$\mathcal{L}_{QD}^P = \sum_j (\alpha_j/2)(2\sigma_{ej} \rho \sigma_{je} - \sigma_{jj} \rho - \rho \sigma_{jj}), \quad (5)$$

is responsible for the incoherent excitation of the QD (e.g. via phonon induced relaxations) from the j -th levels at higher energy that get populated by optical pumping. The α_j are temperature-dependent phonon-assisted scattering rates from the j -th excitonic levels to the state $|e\rangle$ [11]. The transition from the lowest exciton state $|e\rangle$ towards higher energy states can be safely neglected since the experiments of interests are carried out at $KT \ll \omega_j - \omega_x$.

This master equation induces an open hierarchy of dynamical equations. In the SC regime the only meaningful truncation scheme is the one based on the smallness of the excitation density which allows to truncate with respect to the number of photon number-states to be included. The inclusion of one-photon states only gives linear optical Bose-like dynamics. On the other hand, once also 2-photon states are taken into account, saturation and lowest order ($\chi^{(3)}$) nonlinear optical effects arise. Within our model we are able to go beyond 2-photon nonlinear dynamics. We shall show essentially non-perturbative calculations, in the

sense that with respect to the chose pumping intensity, we shall always check the convergence of the presented results to higher-order number-state truncation, see e.g. Fig. 1a.

In the weak excitation regime (linear dynamics) ($\langle\sigma_{ee}\rangle \ll \langle\sigma_{gg}\rangle \simeq 1$), from $\partial_t \langle A \rangle = \text{Tr}(A\dot{\rho})$, we obtain

$$\begin{aligned}\partial_t \langle a \rangle &= -i\tilde{\omega}_a \langle a \rangle - ig \langle \sigma_- \rangle \\ \partial_t \langle \sigma_- \rangle &= -i\tilde{\omega}_x \langle \sigma_- \rangle - ig \langle a \rangle ,\end{aligned}\tag{6}$$

which is a system of two coupled damped oscillators in the absence of any external driving, $\tilde{\omega}_c = \omega_c - i\gamma_c/2$ with $c = a, x$. We also observe that the dephasing rates do not depend on the pumping populations $\langle n_i \rangle$ in contrast to the results of Ref. [7]. We are interested in calculating the steady-state emission spectrum: $S(\omega) = \lim_{t \rightarrow \infty} 2\text{Re} \int_0^\infty \langle a^\dagger(t) a(t+\tau) \rangle e^{i\omega\tau} d\tau$. According to the quantum regression theorem, two-time correlations $\langle A_n(t) A_m(t+\tau) \rangle$ follow the same dynamics of one-body correlation functions $\langle A_m(\tau) \rangle$ but with the one-time correlation $\langle A_n(t) A_m(t) \rangle$ as initial conditions. In our specific case the initial conditions are provided by the steady-state cavity occupation $n_a = \lim_{t \rightarrow \infty} \langle a^\dagger a \rangle$ and by $\mathcal{C} = \lim_{t \rightarrow \infty} \langle a^\dagger \sigma_+ \rangle$. We obtain,

$$n_a = \frac{P_a}{\gamma_a} + \frac{g^2}{\gamma_a} \frac{(\gamma_a + \gamma_x)(\gamma_a P_x + \gamma_x P_a)}{g^2(\gamma_a + \gamma_x)^2 + \gamma_a \gamma_x |\tilde{\omega}_a - \tilde{\omega}_x|^2},\tag{7}$$

$$\mathcal{C} = \frac{g}{\tilde{\omega}_a - \tilde{\omega}_x} (n_a - n_x),\tag{8}$$

where n_x can be obtained from Eq. (7) just exchanging the labels a and x , and $P_x = \sum_i \alpha_i \langle n_i \rangle$. We end up with the following very simple expression, for the PL spectrum

$$S(\omega) = 2\text{Re} \left[\frac{i}{\sqrt{2\pi}} \frac{(\omega - \tilde{\omega}_x) n_a + g\mathcal{C}}{(\omega - R_1)(\omega - R_2)} \right],\tag{9}$$

where the complex polariton energies determining the spectrum resonances are given by

$$R_{(1,2)} = \frac{\tilde{\omega}_a + \tilde{\omega}_x}{2} \mp \frac{1}{2} \sqrt{4g^2 + (\tilde{\omega}_a - \tilde{\omega}_x)^2}.\tag{10}$$

The analytical structure of eq. (9) shows peculiar non-standard features typical of this system, see Fig. 1. Indeed, in addition to the usual dependence in the denominator on the two Rabi frequencies – responsible for the presence of the double-peak structure – we can appreciate a numerator carrying a resonant term proportional to the difference between photon and exciton occupations plus another term proportional to the sole photon steady-state density n_a . Both terms modulate the Rabi resonances with respect to the pumping

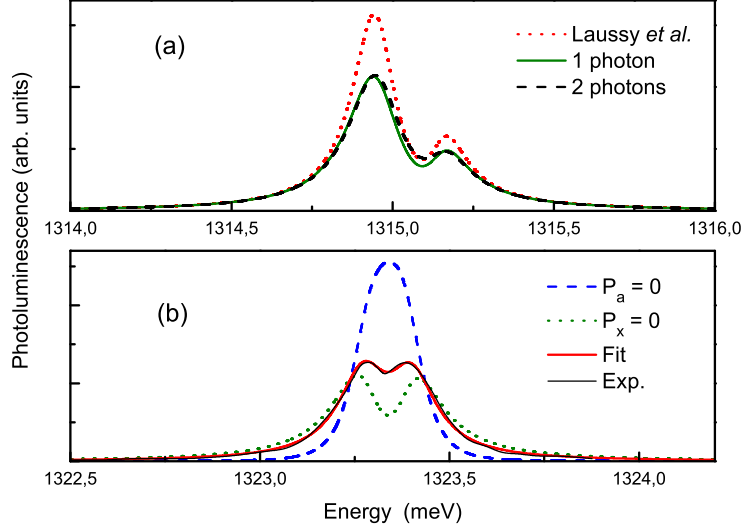


FIG. 1: (Color online). (a): PL spectrum (continuous line) calculated from Eq. (9). We used $\Delta \equiv \omega_a - \omega_x = -0.32$ meV, $g = 90$ μ eV, $\gamma_a = 176$ μ eV, $\gamma_x = 133$ μ eV, $P_a = 0.25 \gamma_a$, $P_x = 5.3 \cdot 10^{-3} \gamma_x$. At these quite low pump intensities the Bose-like spectrum (9) differs only slightly from that calculated including 2-photon states (dashed line). The dotted line (red) emission spectrum calculated according to Ref. [7] differs significantly. (b): PL spectrum (thin continuous line) from the data reported by Reithmaier *et al.* together with the corresponding fit (red). Dashed and dotted lines (blue and green): PL spectra with the same parameters as the fit but with $P_a = 0$ and $P_x = 0$ respectively.

scenario they depend upon. According to Eq. (9) it is not possible that a system in the weak coupling regime enters SC thanks to pumping in contrast to the results of Ref. [7].

Although at low pump intensities, our approach and that of Ref. [7] essentially represent models of a linear Bose-like dynamics of two coupled harmonic oscillators, nontrivial differences can be appreciated. Indeed, Fig. 1a displays the PL spectrum (continuous line) calculated from Eq. (9) compared with the result obtained from the model of Ref. [7] for the same situation (see caption of Fig. 1 for details). At these quite low pump intensities our Bose-like spectrum (9) differs only slightly from that calculated including 2-photon states

(dashed line) and the inclusion of additional photon states does not produce appreciable modifications of the spectrum. On the contrary the emission spectrum calculated according to Ref. [7] differs significantly (dash-dotted line). The difference originates mainly from the effective reduction of the damping rates due to the incoherent pump [7], a feature absent in our theoretical description. The model presented in Ref. [7] reproduces the emission spectra measured by Reithmaier *et al.* [4]. Although the resulting fit is in excellent agreement with the experimental data, some key fitted parameters (e.g. the pumping rates) appears to be in contrast with the experimental results. In particular, as can be inferred from Figs. 3 and 4 of Ref. [4], the measured PL intensities display a very small variation as a function of temperature. On the contrary they estimate pumping rates with a variation larger than a factor four [12]. In addition, Reithmaier *et al.* provides a quite accurate estimate of the mean cavity photon-number (see Additional Information of Ref. [4]) which is one order of magnitude lower than the result of the above mentioned fit. This disagreement strongly supports the absence of any dependence of the damping rates on the pumping rates in agreement with the results of our model. Indeed, all the above experimental features are well reproduced by our master equation (1).

Despite of its analytical simple form, eq. (9) shows a nontrivial dependence of the system on the ratio between the cavity and dot pumping rates. Fig. 1b displays one PL spectrum (thin continuous line) from the data reported by Reithmaier *et al.* together with the corresponding fit we obtain using Eq. (9) (thicker continuous line). Beside the nearly coincidence of the two curves in this situation, Eq. (9) is also in very good agreement with the experimental spectra of Ref. [4] at different detunings. As absolute information on the PL intensities is commonly not available from this kind of experiments, we cannot obtain absolute values for the pumping rates. Instead we gather from the fit the ratio $P_a/P_x = 0.86$. We also obtain $g = 76$ meV, $\gamma_a = 100$ μ eV, $\gamma_x = 35$ μ eV. In order to evidence the impact that the pumping mechanism can have on SC emission lines even at very small detuning, we plotted in Fig. 1b two spectra with the same parameters as the fit but with $P_x = 0$ (dotted line) and $P_a = 0$ (dashed line). These plots show that, close to the SC threshold, incoherent pumping of the exciton level can even hinder the line splitting. This is the case of the system investigated by Reithmaier *et al.* [4]. This result put forward the importance of addressing the correct mixed quantum steady state in order to describe the experimental results.

Another situation for a similar system has been recently reported in Ref. [2]. Some first

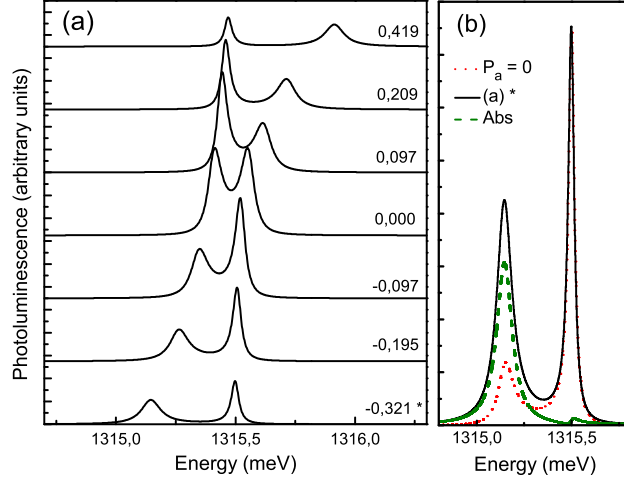


FIG. 2: Color online. (a) PL spectra calculated from Eq. (9 at different detunings reproducing the experimental observation of the SC of a QD-cavity system reported in Ref. [2]). (b) continuous line: one spectrum of panel a (the one with the lowest cavity energy) compared with the corresponding spectrum obtained using $P_a = 0$ (dotted line) and with the corresponding absorption spectrum (dashed line).

results based on the present multi-photon model (1) are depicted in Fig. 2a. It displays emission spectra at different detunings reproducing one experimental observation of the SC of a QD-cavity system reported in Ref. [2]. Although the parameters have been chosen in order to fit just one of the spectra, our model provides results which agree very well [13] with all the spectra presented in Fig. (2b) of Ref. [2] changing only the cavity energy. We can thus unravel the ratio between the cavity-mode and the exciton pumping in this kind of experiments. In this case we obtain $P_a/P_b = 0.13$. Fig. 2b clearly underlines the importance of taking into account the correct quantum steady state resulting from the interplay of pumping and decay (of both the cavity mode and the exciton level) in order to describe properly the SC experiments. It shows one spectrum of Fig. 2a ($\Delta = 0.1$ meV) (continuous line) together with the emission spectrum obtained neglecting the pumping P_a of the cavity mode and the absorption spectrum under a coherent pump. As can be clearly seen without the proper P_a pumping mechanism, the calculated results differ significantly

from the experiment.

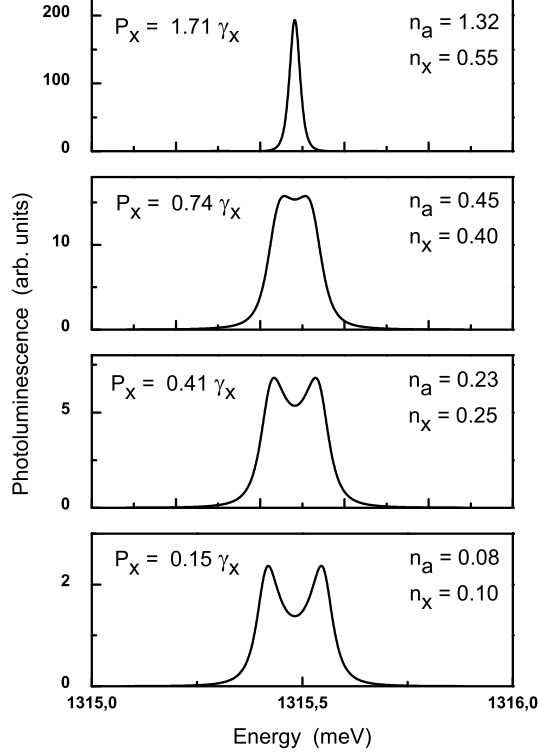


FIG. 3: Transition from Rabi splitting to single-emitter laser when increasing the pumping rate.

Our model is also well suited to describe situations well beyond that of linear or lowest order nonlinear effects. Fig. 3 displays emission spectra obtained at different pumping intensities. These results have been obtained after an exact calculation of the SC quantum dynamics achieved truncating the photon number-states only when including larger numbers (and hence dealing with a larger system of equations of motion) does not produce any change in the emission spectrum. We used the same broadenings of Fig. 2, a coupling $g = 76 \mu\text{eV}$ and we set $P_a = 0$ and $\Delta = 0$. Increasing the excitation a reduction of the Rabi splitting due to saturation effects is clearly observable. The upper panel show that as soon as a small popoulation inversion ($n_x > 0.5$) is reached an important narrowing of the linewidth appears indicating a thresholdless lasing behaviour. It seems worth underlining that, within our model, both the linewidth narrowing and the build up of lasing are purely fermionic effects not to be confused with the narrowing due to the incoherent pumping found in the

model of Ref. [7] (see also [5]).

In conclusion, we have given a simple theoretical framework with a reliable predicting character for the analysis of photoluminescence properties under incoherent excitation of single quantum dot microcavity devices in a steady state maintained by a continuous incoherent pumping. On the contrary to currently exploited approaches in the literature, we constructed a completely positive quantum master equation able to provide physically sensible results free from artifacts due to the reduced phenomenological description. Remarkably, our theoretical framework represents a simple and flexible tool naturally suitable for the analysis of the impact of the interplay between the two feeding mechanisms (i.e. P_a and P_x) onto the quantum statistic of the emitted quanta at different excitation intensities (under current development). At low pumping rates we obtained an analytical expression for the emission spectrum which directly shows the very different impact of the two feeding mechanisms on the spectra. Our model showed a great flexibility proposing simple but sensible descriptions of various experiments in the strong-coupling regime. It provides essentially non-perturbative results which can be exploited for the efficient modeling of future SC optoelectronic devices.

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